One-way multigrid method in electronic structure calculations

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Abstract

We propose a simple and efficient one-way multigrid method for self-consistent electronic structure calculations based on iterative diagonalization. Total energy calculations are performed on several different levels of grids starting from the coarsest grid, with wave functions transferred to each finer level. The only changes compared to a single grid calculation are interpolation and orthonormalization steps outside the original total energy calculation and required only for transferring between grids. This feature results in a minimal amount of code change, and enables us to employ a sophisticated interpolation method and noninteger ratio of grid spacings. Calculations employing a preconditioned conjugate gradient method are presented for two examples, a quantum dot and a charged molecular system. Use of three grid levels with grid spacings $2h$, $1.5h$, and h decreases the computer time by about a factor of 5 compared to single level calculations.

71.15.-m, 71.15.Nc, 71.15.Mb, 71.15.Pd

I. INTRODUCTION

Recently, the usefulness of the real-space technique based on three-dimensional uniform grid and higher-order finite-difference formula[1] has been demonstrated[2,3] for the electronic structure calculations within the framework of the Kohn-Sham (KS) density functional theory (DFT).[4] All the computations are performed in real space without resort to fast Fourier transforms as in the planewave formalism. The major parts of calculations are local operations, so the algorithm is easily parallelized. Furthermore, explicit storage of the Hamiltonian matrix elements can be avoided, since the Laplacians and potential-wave functions multiplications are respectively evaluated by the finite-difference operation on the wave functions and a simple one-dimensional vector multiplications.

Since the number of grids N_g is order of $10^5 \sim 10^6$, which increases further with the system size and/or the level of accuracy, one requires an efficient numerical procedure for the Hamiltonian diagonalization in finite-difference real-space schemes. Iterative diagonalization methods are usually employed as in other modern electronic structure calculations, and due to the orthogonality condition between the KS orbitals the complexity of this iterative diagonalization scales as $O(N_b^2 N_g)$, where N_b represents the number of lowest states taken into account. It turns out that the prefactor of this scaling can be very dependent on the details of calculation scheme, and the development of a new algorithm which results in the optimal prefactor is a very important and challenging problem at the moment. Among the most promising approaches in the literature are the multigrid (MG) algorithms.[5,6] MG methods originated as attempts to accelerate relaxation methods and they have been very successful in improving the speed and accuracy in a wide variety of science and engineering applications by combining computations at different scales of grid levels. In the context of DFT electronic structure calculations, several groups have already applied the multigrid scheme to the solution of KS equations and demonstrated its efficiency: Briggs *et al.* adopted coarse-grid-correction multigrid algorithm to the calculations of various periodic and nonperiodic systems.[7] Ancilotto *et al.* implemented full multigrid diagonalization procedure to study the fragmentation of charged Li clusters.[8] While these two works employed the pseudopotentials, Beck *et al.* has demonstrated the feasibility of all-electron grid calculation by employing full multigrid algorithm.[9] These authors typically use integer ratio of grid spacings (e.g., $4h$, $2h$, and h) and correction multilevel algorithm (V-cycle).

In this article, we introduce a simple one-way multigrid algorithm[5] to accelerate self-consistent electronic structure calculations based on iterative diagonalization. Calculations start from the coarsest grid level and approximate solutions are transferred successively up to the finest grid. An interesting aspect of this method is that the number of interpolation is minimized : Interpolations are performed outside of original total energy calculation part, hence only for (number of grid levels -1) times when the wave functions are transferred to the next finer grid level. It enables us to use an accurate interpolation scheme and the noninteger ratio of grid spacing in the hierarchy of grids. Specifically we employ three different uniform grids spacings, $2h$, $1.5h$, and h to obtain the solution at the resolution of grid spacing h, in which calculations on the preceding two coarse grids provide a good initial guess of the wave functions for the finest level calculation. We demonstrate the efficiency of the current scheme on the twenty-electron quantum dots and the charged H cluster systems in which the ionic potentials have been replaced by *ab initio* pseudopotentials. The comparison with

a single-level calculations shows a factor of 5 improvement in CPU time.

II. METHODS

A. Basic issues

The iterative total energy minimization based on DFT is a nonlinear problem in which the KS equations (Hartree atomic units are used throughout this paper)

$$\left\{-\frac{1}{2}\vec{\nabla}^2 + V_{ext}(\vec{r}) + \tilde{V}_H(\vec{r}) + \tilde{V}^{\sigma}_{xc}(\vec{r})\right\}\psi^{\sigma}_j(\vec{r}) = \epsilon^{\sigma}_j\psi^{\sigma}_j(\vec{r}),\ \sigma =\uparrow,\downarrow, j = 1,\cdots,N_b \tag{1}$$

and the Poisson equation

$$\vec{\nabla}^2\tilde{V}_H(\vec{r}) = -4\pi\rho(\vec{r}), \tag{2}$$

are closely coupled in the self-consistency loop.[10] Here $\tilde{V}_H(\vec{r})$ and $\tilde{V}^{\sigma}_{xc}(\vec{r})$ respectively represent the input Hartree and spin dependent exchange-correlation potential, at each iteration within the self-consistent calculations. $V_{ext}(\vec{r})$ stands for the external potential, and the charge density $\rho(\vec{r})$ is defined as the squared summation of the occupied KS orbitals. In the higher-order finite-difference real space formulation, the KS and Poisson equations are discretized on a uniform grid. The Laplacian operation is evaluated by the higher-order finite difference formula[1] which is characterized by the finite-difference order N and grid spacing h:

$$\frac{d^2}{dx^2}f(x) = \sum_{j=-N}^{N} C_j f(x+jh) + O(h^{2N+2}), \tag{3}$$

where $\{C_j\}$ are constants.

In the present work solutions of the KS equations for the lowest N_b eigenstates are found by the iterative preconditioned conjugate gradient method of Bylander, Kleinman, and Lee,[11,12] for a given total potential $V^{\sigma}_{KS}(\vec{r}) = V_{ext}(\vec{r}) + \tilde{V}_H(\vec{r}) + \tilde{V}^{\sigma}_{xc}(\vec{r})$. The Hartree potential $V_H(\vec{r})$ is obtained by solving the Poisson equation. Note that, for each self-consistent step, we need to solve two Poisson equations for the given input and output charge densities. For the finite systems considered here the boundary values of Hartree potential are evaluated using a multipole expansion of the potential of the charge distribution and the relaxation vectors at the boundary are set to zero for the Dirichlet boundary conditions. The solution of Poisson equation inside of the box has been first generated by a Fourier method with low order finite difference ($N = 1$),[6] and it has been subsequently relaxed by the preconditioned conjugate gradient method[13,14] with higher-order finite difference formula. At each step we choose the new input density and potential using a simple linear mixing of output and input densities.[11]

After obtaining orbitals and density from self-consistent solutions of Poisson and KS equations, the total electronic energy is obtained:

$$E_{tot} = \sum_{\sigma,j}\epsilon^{\sigma}_j - \sum_{\sigma}\int d^3r\{\tilde{V}_H(\vec{r}) + \tilde{V}^{\sigma}_{xc}(\vec{r})\}\rho^{\sigma}(\vec{r}) + \frac{1}{2}\int d^3r'\int d^3r\frac{\rho(\vec{r})\rho(\vec{r}')}{\mid \vec{r}-\vec{r}' \mid} + E_{xc}[\rho^{\uparrow}(\vec{r}),\rho^{\downarrow}(\vec{r})], \tag{4}$$

where the summations over the single particle energy (ϵ_j^σ) are carried out for all the states below the Fermi level, and $E_{xc}[\rho^\uparrow(\vec{r}), \rho^\downarrow(\vec{r})]$ is the exchange-correlation energy. For our local spin density approximation we use the Perdew and Zunger's parameterization of the Ceperley and Alder's quantum Monte Carlo data.[15]

B. One-way multigrid method

The most time-consuming part of the self-consistent electronic structure calculations described in the previous subsection is the iterative solution of KS equations. The sources of this computation bottleneck can be traced to broadly two (but closely related) aspects of self-consistent iterative diagonalization schemes. First of all, in general we do not have a good initial guess of wave functions, which generate density, and hence $\tilde{V}_H(\vec{r})$ and $\tilde{V}_{xc}^\sigma(\vec{r})$ in Eq. (1). So initial several self-consistency steps will be used to obtain solutions of biased Hamiltonians, although they tend to be the most time-consuming part. Secondly, in single iterative solution of KS equations, a direct application of a relaxation method on the fine grid has trouble in damping out the long-ranged or slowly varying error components in the orbitals. This can be understood by the usual spectral analysis of relaxation scheme, or considering that the nonlocal Laplacian operation on a fine grid is physically short-ranged. This means that there is an imbalance in the relaxation step for the long-ranged and short-ranged error components.

MG is a quite general concept, and the choice of a specific algorithm is very dependent on the problem under consideration. For our purpose, we seek a procedure which generates a good initial guess for the finest grid calculation and effectively removes long-range error components of wave functions in the solution of KS equations. In this work, we employed the one-way multigrid scheme with three different uniform grids with noninteger ratio of spacings, $2h$, $1.5h$, and h. The calculation starts from the coarsest grid $2h$, and in each grid-level calculation, Eqs. (2) and (1) are solved self-consistently as in the usual single-level algorithm. After each self-consistent calculation on a coarse grid, only wave functions are interpolated to the next fine grid, and another set of self-consistent calculation is performed. Since that the interpolated wave functions usually do not satisfy the orthonormality condition any more, we take an extra Gram-Schmidt orthogonalization process after each orbital interpolation. So we have two interpolations and two Gram-Schmidt orthogonalization processes for our hierarchy of three grid systems. In Fig. 1, we summarize the algorithmic flow of the procedures.

While an efficient interpolation/projection scheme is a crucial ingredient of any successful application of MG method, we note that it can be also time-consuming and tricky part due to the physical conditions such as orthonormality of wave functions. Hence our strategy, which is the characteristic of the current scheme, is to minimize the number of data transfer between different grid levels, while employ a sophisticated interpolation method which is very accurate and allow us to use a noninteger ratio of grid spacings. Specifically, we used a three-dimensional piecewise polynomial interpolation with a tensor product of one-dimensional B-splines as the interpolating function.[6,16] A piecewise cubic polynomials have been taken as B-splines.

III. EFFICIENCY AND DISCUSSIONS

We consider two different electronic systems of a localized quantum dot model and a charged hydrogen cluster to demonstrate the efficiency of the present algorithm. We first take a twenty-electron quasi two-dimensional quantum dot modeled by an anisotropic parabolic confinement potential[3] $V_{ext}(\vec{r}) = \frac{1}{2}\omega_x^2 x^2 + \frac{1}{2}\omega_y^2 y^2 + \frac{1}{2}\omega_z^2 z^2$. In-plane potential is characterized by the confinement energies $\omega_x = \omega_y = 5$ meV, and $\omega_z = 45$ meV has been taken to reproduce the dot-growth z-direction confinement caused by the quantum wells and heterojunctions.[3] Our calculations for anisotropic parabolic dot in GaAs host material (dielectric constant $\epsilon = 12.9$, effective mass $m^* = 0.067 m_e$) are based on the effective mass approximation, and rescaled length and energy units are $a_B^* = 101.88$ Å and 10.96 meV, respectively. Uniform grid spacing $h = 0.3 a_B^*$ with box size $81 \times 81 \times 21 {a_B^*}^3$ have been used, hence the number of grid points is about 1.4×10^5 points at the finest grid level ($h = 0.3 a_B^*$) while only about 1.6×10^4 points at the coarsest grid level ($2h = 0.6 a_B^*$). Finite difference order $N = 3$ has been used at grid levels h and $1.5h$, while $N = 1$ for grid level $2h$, to solve a set of spin-polarized KS equations with fifteen orbitals in each spin channel. Noninteracting eigenstates (Hermite polynomials) are used as an initial guess for the coarsest grid calculation.

The CPU times for each self-consistent iteration are shown in Fig. 2. The horizontal axis stands for the self-consistency iteration index, while the vertical axis is the required computer time for a given iteration step. The case of the present three-level one-way multigrid algorithm is shown in the lowest panel (c). Comparing with the results of a single-level calculation shown in the panel (a), we see significant savings of the computation time, in which total computation time is about 5 times shorter than a single-level calculation. While the three-level MG scheme requires more number of self-consistent iterations (28 iterations compared with 20 iterations), they are mostly performed in the coarsest grid $2h$, and at the finest grid level h we only need several iterations. Interpolation and orthonormalization steps are indicated by downward arrows, which take only a small amount of computation time.

To further demonstrate the advantage of the usage of the intermediate grid spacing $1.5h$ in our three-level scheme, we show the CPU time of two-level ($2h$ and h) calculation in the panel (b). While the number of iterations taken in the finest grid h is still much smaller than the single-level calculation, it is much larger than that in the three-level calculation, resulting in the ratio of computation times $5 : 2 : 1$ for one-, two-, and three-level grid calculations. Although noninteger ratio of grid spacing is not widely used in MG applications, this clearly shows its usefulness.

We obtained similar results of factor 5 improvements in computation speed for other test cases, such as the *ab initio* nonlocal pseudopotential calculation of a charged hydrogen cluster H_9^+. Details of the calculation are identical to those of quantum dot calculations, except that ionic external potentials are treated by separable[17] nonlocal pseudopotential generated by the method of Troullier and Martins.[18] Finite difference order $N = 6$ at grid h and $1.5h$, and $N = 1$ for the grid $2h$ have been used to solve spin-unpolarized KS equations with the lowest 6 states. The number of grid points involved in the finest grid calculation is 3.5×10^5, while it is 4.3×10^4 for the coarsest grid calculation.

We have to emphasize that the improvements seen in previous examples are purely induced by the a simple usage of MG idea, in which the only modification from the original

single-level code was the addition of an outer loop which transfer the wave functions. We can expect that the introduction of the MG scheme at different stages of calculations, such as the correction path for the relaxation of KS orbital or Hartree potential, will result in further improvements. To do so, we will need additional residual computation and projection steps that can be combined with our conjugate gradient solvers. We also note that it will require an interpolation strategy and grid levels which are different from the current method. Finally, we point out that this type of one-way multigrid idea is very similar to often-used practices in plane-wave calculations based on iterative diagonalization, in which a solution is first found at one energy cutoff (equivalent to a coarse grid) and used as the input to a higher energy cutoff calculation (equivalent to a finer grid). This corresponds to interpolating solutions from a coarser to a finer grid using Fourier components.

IV. CONCLUSIONS

In this work, we demonstrated that the introduction of a simple one-way multigrid method greatly improves the efficiency of real-space electronic structure calculations based on the iterative solution of KS equations. While minimizing the number of data transfer between grids, we employed an accurate interpolation method, which enabled us to incorporate three-level grids with noninteger ratio of grid spacings. Our general strategy of using $2h$, $1.5h$, and h, showed a factor 5 improvement of computation time, while it required only minimal computer code modifications. The usefulness of the intermediate grid step $1.5h$ has been shown by comparing the current scheme with two-level ($2h$ and h) calculations.

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FIGURES

FIG. 1. Schematic diagram of the present one-way multigrid algorithm discussed in the text. The self-consistent calculation at each level is done by using preconditioned conjugate gradient relaxation. The values in circles, $2h$, $1.5h$, and h stand for the uniform grid spacing for a given level. The calculation starts at the coarsest level (level 1, $2h$) at the bottom, and ends at the finest grid (level 3, h) at the top. Orbital interpolation and orthogonalization step is taken after each coarse grid (level 1 and 2) calculation.

FIG. 2. CPU time vs. self-consistent iteration number for 20-electron quantum dot calculations in (a) single-level (h), (b) two-level ($2h$ and h), and (c) three-level ($2h$, $1.5h$, and h) schemes. Within the local spin density approximation, we minimized the total energy with respect to the electronic degree of freedom. Downward arrows in (b) and (c) indicate the interpolation-orthogonalization steps. Calculations are performed on a DEC alpha 433au personal workstation.

level 3
level 2
level 1
h
1.5h
2h
self-consistent calculation
interpolation & orthogonalization
self-consistent calculation
interpolation & orthogonalization
self-consistent calculation
Computation flows from left to right

Fig. 1

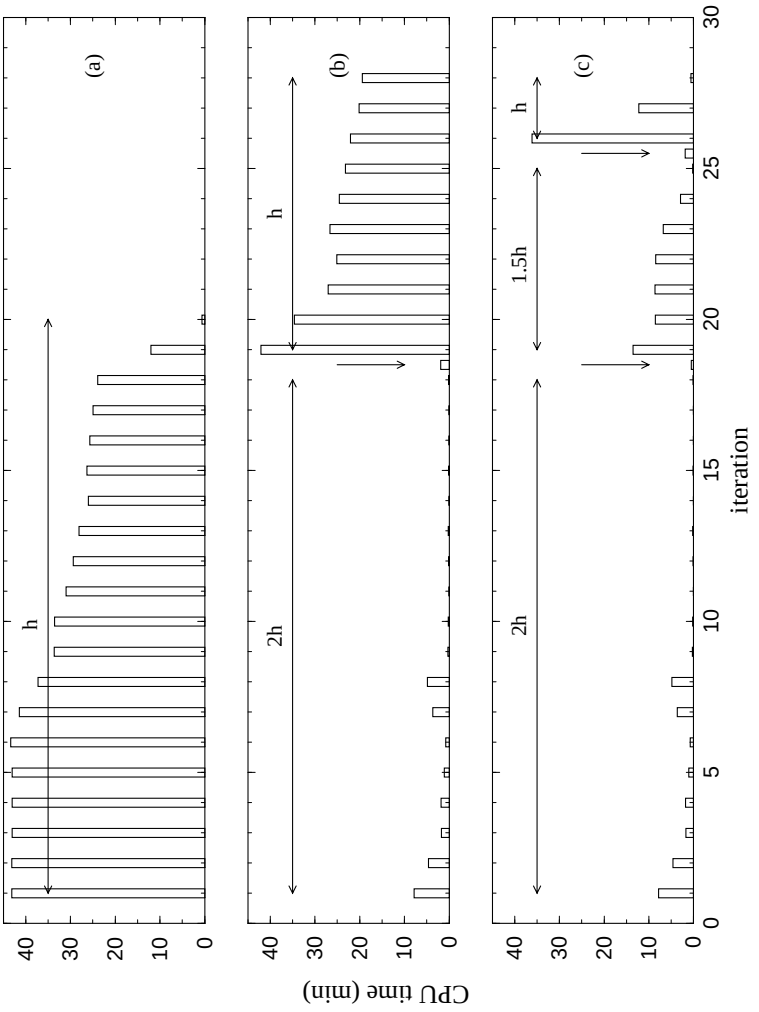

Fig. 2